

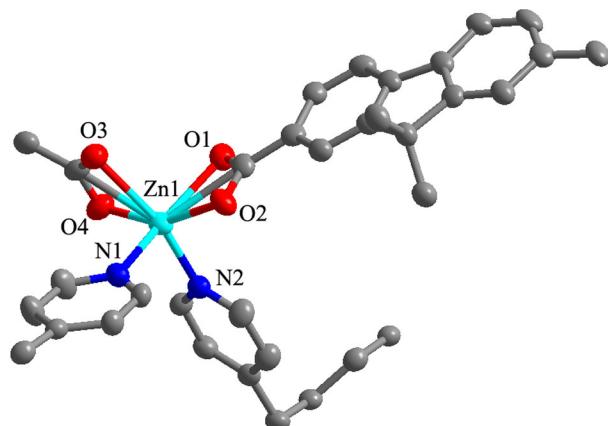
[Supporting information]

## Versatile Entangled Metal-Organic Frameworks Modulated by N-Donor Ligands

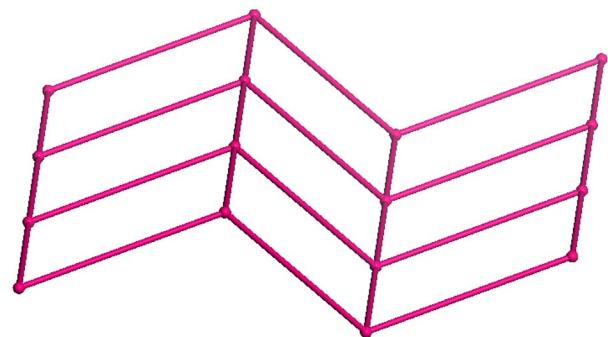
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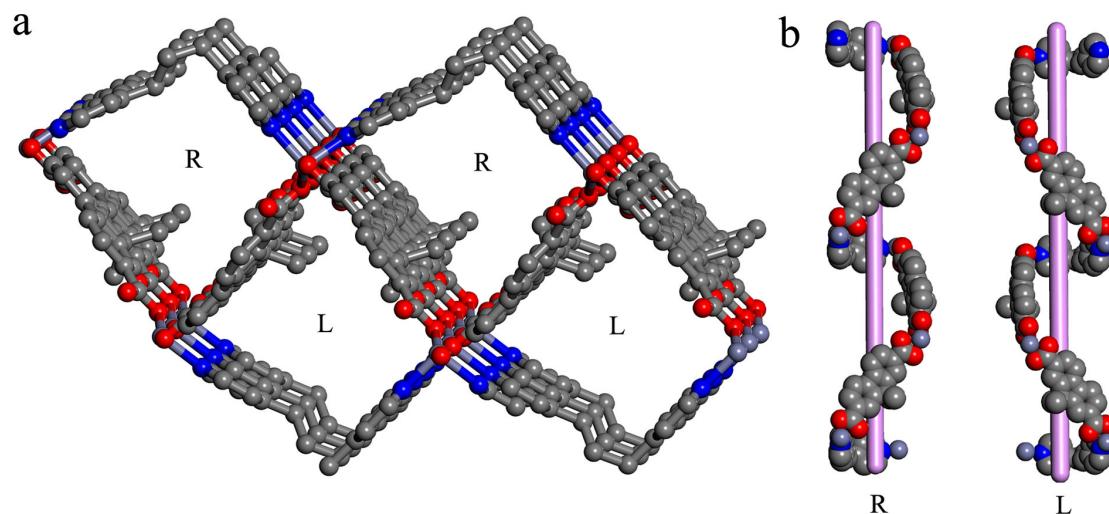
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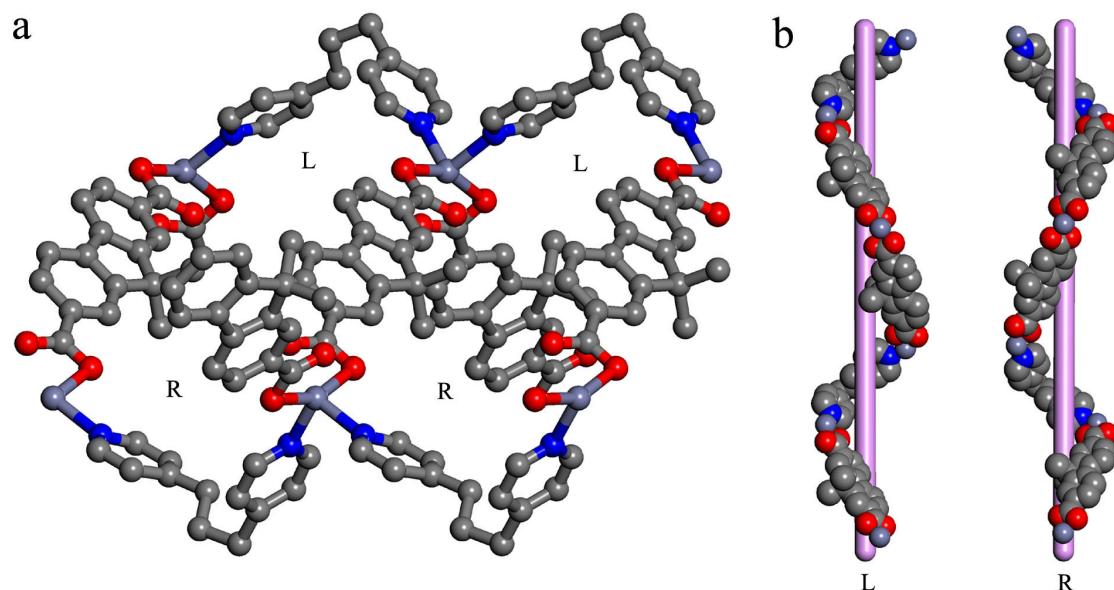
**Fig S1.** Molecular structure of **1** showing the coordination environment of the  $\text{Zn}^{2+}$  ions Hydrogen atoms are omitted for clarity.



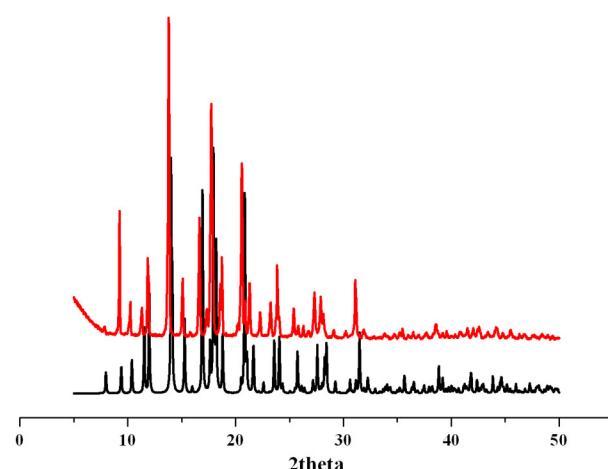
**Fig S2.** View of the topology of 2D (4, 4) net.



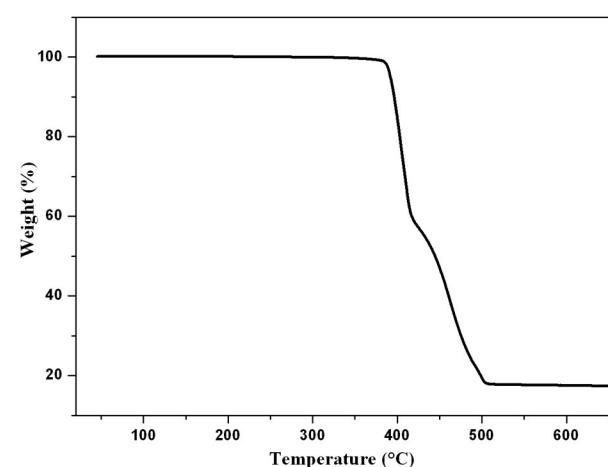
**Fig. S3.** (a). View of double-layered sheet assembled by **A** helices. (b). View of the **A** helice.



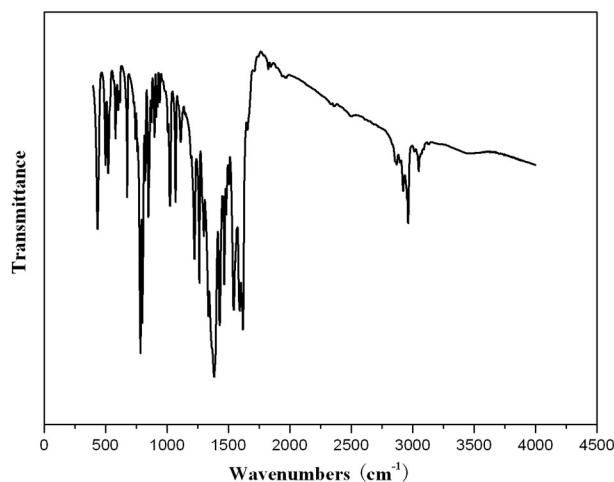
**Fig. S4.** (a). View of double-layered sheet assembled by **B** helices. (b). View of the **B** helice.



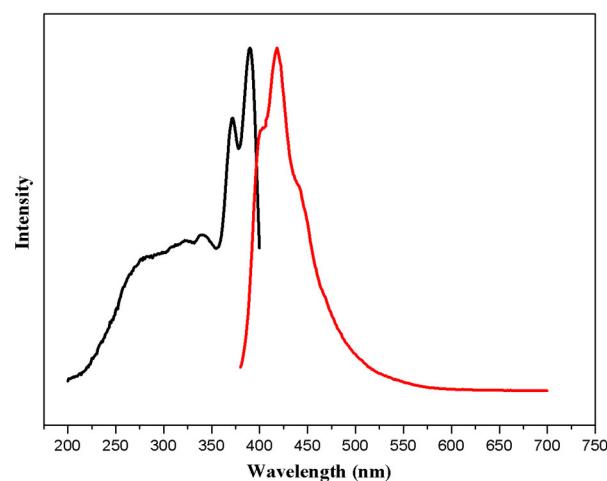
**Figure S5.** The simulated (black) and experimental (red) XRPD patterns for **1**.



**Figure S6.** TGA curve of **1**.



**Figure S7.** IR (4000–400  $\text{cm}^{-1}$ ) spectrum of the compound **1**.



**Fig. S8.** Excitation (black) and emission (red) spectra of **1**. Excitation at 365 nm leads to strong blue fluorescent emission band at 417 nm, which can probably be assigned to intraligand ( $\pi-\pi^*$ ) fluorescent emission because almost similar emissions are observed for the free H<sub>2</sub>MFDA.

Table S1. Selected bond distances ( $\text{\AA}$ ) and angles ( $^\circ$ ) for **1**.

N(1)-Zn(1)	2.114(5)
N(2)-Zn(1)	2.078(4)
O(1)-Zn(1)	2.280(4)
O(2)-Zn(1)	2.082(4)
O(3)-Zn(1)	2.341(4)
O(4)-Zn(1)	2.056(5)
O(4)-Zn(1)-N(2)	97.63(18)
O(4)-Zn(1)-O(2)	154.15(17)
N(2)-Zn(1)-O(2)	100.00(17)
O(4)-Zn(1)-N(1)	100.56(18)
N(2)-Zn(1)-N(1)	95.67(18)
O(2)-Zn(1)-N(1)	96.36(18)
O(4)-Zn(1)-O(1)	99.22(17)
N(2)-Zn(1)-O(1)	96.00(16)
O(2)-Zn(1)-O(1)	60.30(16)
N(1)-Zn(1)-O(1)	155.42(17)
O(4)-Zn(1)-O(3)	59.69(18)
N(2)-Zn(1)-O(3)	157.19(18)
O(2)-Zn(1)-O(3)	102.21(17)
N(1)-Zn(1)-O(3)	87.00(17)
O(1)-Zn(1)-O(3)	90.53(16)